



Docket No.: C070973/0231937

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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| In re Application of: R. A. Van Benthem |) | Examiner: John D. Freeman |
| |) | |
| Serial No.: 10/560,212 |) | |
| |) | |
| Filed: December 9, 2005 |) | Art Unit: 1794 |
| |) | |
| |) | |
| For: ENCAPSULATED MATERIALS |) | |
| |) | |

DECLARATION OF RUDOLFUS VAN BENTHEM, Ph.D.,
UNDER 37 C.F.R. § 1.132

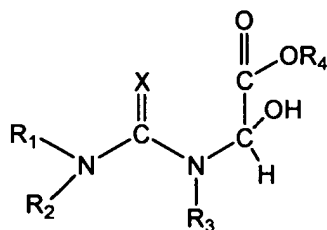
Sir:

I, Rudolfus van Benthem, a citizen of the Netherlands, declare as follows:

1. I am the inventor of the above-mentioned application.
2. I received a Masters' degree in Organic Chemistry from Leiden University, the Netherlands, in 1991. I received a Ph.D. in Organic Chemistry from the University of Amsterdam, the Netherlands, in 1995.
3. I was employed by DSM in 1995 as a scientist in resins chemistry. I became Research Group leader in 1998, and was appointed Competence Manager Network Chemistry in 2002. In this capacity I am responsible within DSM (world-wide) for the knowledge regarding crosslinking of crosslinked materials, chemistry of thermosetting materials such as adhesives (including melamine resins), coatings, composites and elastomers. From 2002 to the present time, I have been a part-time Professor of Coatings Technology at the Eindhoven University of Technology.

4. The present application discloses and claims a process for forming capsules comprising the steps of: (1) forming a solution of an amino compound (V) from a mixture of an amino compound and an alkanol hemiacetal in a solvent; (2) forming a dispersion of a core material in the solution; (3) depositing amino compound (V) as a resin upon the surface of the core material to form capsules; and (4) optionally hardening and/or recovering the capsules, whereby steps (1) and (2) are executed in either order or simultaneously, and wherein amino compound (V) has the following formula

(V)



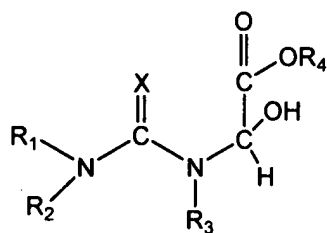
where: X is NR₅; R₄ is equal to a C₁-C₁₂ alkyl group, aryl group, aralkyl group or cycloalkyl group; R₁, R₂, R₃, R₅ are equal to an H, alkyl, cycloalkyl, aryl or heterocyclic group; and R₁, R₂, and R₅ or R₁, R₂, and R₃ may together form a heterocyclic group. The specification discloses that “[a]n aim of the present invention is to provide a process for forming capsules, which do not release free formaldehyde.”¹ (Paragraph 9, lines 1-3.) The specification also discloses the preparation of capsules in accordance with the claimed invention; Example 1 discloses use of melamine with methyl glyoxylate and

¹ Citations to the present application refer to the published application, U.S. 2007/0092726.

Examples 2 and 3 disclose use of melamine with methanol hemiacetal. (Paragraphs 53-55; see also paragraph 36.)

5. I am aware that a Final Office Action has issued with regard to the present application. It is my understanding that in the Office Action, the Examiner asserted that the claimed process for forming capsules is not patentably distinct from Rätzsch, WO 02/48261 ("Rätzsch ") in view of Skoultchi, U.S. Patent No. 4,770,668 ("Skoultchi"). The Examiner has relied on these documents as disclosing and/or suggesting a process for forming capsules comprising the steps of: (1) forming a solution of an amino compound (V) from a mixture of an amino compound and an alkanol hemiacetal in a solvent; (2) forming a dispersion of a core material in the solution; (3) depositing amino compound (V) as a resin upon the surface of the core material to form capsules; and (4) optionally hardening and/or recovering the capsules, whereby steps (1) and (2) are executed in either order or simultaneously, and wherein amino compound (V) has the following formula

(V)



where: X is NR₅; R₄ is equal to a C₁-C₁₂ alkyl group, aryl group, aralkyl group or cycloalkyl group; R₁, R₂, R₃, R₅ are equal to an H, alkyl, cycloalkyl, aryl or heterocyclic group; and R₁, R₂, and R₅ or R₁, R₂, and R₃ may together form a heterocyclic group. (Paper No. 20100106 at 2-7.)

6. I am familiar with Rätzsch which is cited in the pending Final Office Action. (Paper No. 20100106 at 2-7.) Rätzsch discloses a method for curing aminoplast resins in which inorganic particles, which have a laminated structure and which comprise interlamellary exchangeable cations of the following type: alkali cations, alkaline-earth cations, aluminum cations, iron cations and/or manganese cations, are used as curing agents. (Rätzsch, Abstract, lines 1-6.) The resins include polycondensates of melamine or melamine derivatives and C₁₋₁₀-aldehydes. (Paragraph 14, lines 1-2.) Rätzsch also discloses that “[t]he invention also relates to aminoplast resins cured in such a manner, to [provide] semi-finished products and ... molding materials.” (Abstract, lines 6-8.) Among the disclosed “semifinished products and moldings” disclosed by Rätzsch are microcapsules (Paragraph 12, lines 1-5, Paragraphs 41, 44, 60, and Paragraph 67, lines 1-2 and 6-7), yet no Examples are disclosed of the preparation of microcapsules. (Examples 1 and 2 disclose the preparation of impregnated sheet-like substrate materials, and Example 3 discloses the preparation of a laminate.)

7. I am also familiar with Skoultchi which is cited in the pending Final Office Action. (Paper No. 20100106 at 2, 5-6.) Skoultchi discloses “adducts of cyclic ethylene urea which are useful as permanent press agents. These adducts include acid, polyacid ester, and multiester derivatives of cyclic ethylene urea and can be produced by the reaction of the cyclic ethylene urea with glyoxylic acid and glyoxylic acid derivatives, specifically ester acetals of glyoxylic acid. Such materials impart a high degree of permanent press properties to cellulose and cellulose/polymer blend fabrics.” (Skoultchi, Abstract.) Skoultchi discloses that “[t]he compounds ... can be

prepared by the reaction of a 2-imidazoline derivatives [sic] with glyoxylic acid or an ester acetal of glyoxylic acid to form a monoester or polyester of imidazolidinone.” (Col. 2, lines 63-66.)

8. It is my understanding that in the Final Office Action, the Examiner asserted that “[a]t the time of the invention, it would have been obvious to one of ordinary skill in the art to use glyoxylic acid hemiacetals [as disclosed by Skoultchi] as the aldehyde of Rätzsch’s invention because it was a known functional equivalent to Rätzsch’s exemplary aldehydes while avoiding the use of toxic formaldehyde, yet providing reactivity over a wide pH range.” (Paper No. 20100106 at 2.)

9. The disclosure of preparation of adducts for use as permanent press agents in Skoultchi would not have provided any expectation of success in preparing capsules, as in the claimed process. In Skoultchi a solution of the adduct material is applied to a fabric and treated to crosslink the fabric. (Col. 5, line 63 – Col. 6, line 24.) Skoultchi provides no suggestion as to whether the disclosed adducts, if present in a dispersion containing a core material and a wall forming materials, i.e., the adduct, would phase separate from the continuous phase so as to result in encapsulation of the core material.

10. Furthermore, Skoultchi discloses the use of “adducts of cyclic ethylene urea.” The reactant used by Skoultchi is 2-imidazolidinone, i.e., ethylene urea, which does not encompass or suggest melamine or melamine derivatives for use in the claimed process of forming capsules. The urea compounds of Skoultchi have an oxygen molecule in the corresponding position as substituent X in formula (V) which is NR_5 in the claimed process. Accordingly, the 2-imidazolidinone and melamine differ in

the number and position of amino groups. It is known that ethylene ureas that form resins with formaldehyde have different reactivities than melamine-formaldehyde resins. It would not have been expected that these resins would work in the same way. One could not predict success from the use of urea resins as in Skoultchi for use as permanent press agents in using melamine resins in a process for encapsulation.

11. In addition, one skilled in the art would not have predicted success in using methyl glyoxylate as the aldehyde in place of the aldehyde of Rätzsch reacted with melamine, in the claimed process for preparing capsules. To demonstrate that the claimed process is not obvious over Rätzsch in view of Skoultchi, the results of studies performed that are relevant to this issue are provided. These studies were performed with my knowledge by a scientist within my Research Group at DSM. Although the evaluation was not performed under my direct supervision and direction, I have responsibility as the Research Group leader for the results obtained which are presented as follows.

Evaluation

12. An evaluation was made of the performance of glyoxylic acid, which is an aldehyde having an acid end group, in attempting to prepare capsules by reacting melamine with glyoxylic acid. Glyoxylic acid was used in place of methyl glyoxylate, the methyl ester acetal of glyoxylic acid, in the procedure disclosed in Example 1 of the present specification. Glyoxylic acid was used in the same and in a similar molar ratio as was methyl glyoxylate in Example 1.

13. When melamine was reacted with methyl glyoxylate in accordance with Example 1 of the present specification, capsules resulted. Example 1 is duplicated

below and additionally includes, by way of a footnote, the molar ratio of melamine to methyl glyoxylate used in the procedure.

Example I of the Present Specification:

In a reaction vessel 62.9 gram melamine, 89.2 gram methyl glyoxylate and 64.8 gram water was added.^[2] This mixture was under constant stirring, heated up in [an] oil [bath] of 80° C. until the melamine reacted completely with the glyoxylate and the resin is clear. Then 50 gram of paraffin oil at 80° C., in which 5 mg of a non-water soluble dye (Solvent Blue 59) is, under fierce stirring with a Ultra Turrax T25 at 24,000 rpm, carefully mixed in. To stabilize the dispersion of oil and resin a surfactant (Disperbyk-181) was added. The warm oil/resin dispersion was carefully, under stirring, added to cold water; in this fashion, hardening of the capsules was achieved through cooling. The oil/resin dispersion precipitates in the water. The precipitate was filtered out with a paper filter, leaving a colourless liquid as permeate; the light blue powder was dried in vacuum oven at 50° C. The powder was washed with heptane to determine the amount of "free" paraffin oil. The measured weight loss was 3.6%. The washed powder, still light blue in colour, was then [analyzed] with Differential Scanning Calorimetry and showed a glass [transition] at [-]70° C.^[3], showing that the powder contained paraffin oil. Crushing the powder in a mortar afforded a more intense blue coloured smearing paste. [Paragraph 53.]

14. In the evaluation of glyoxylic acid, four attempts were made to prepare capsules which are identified as Attempts A-D below. In the procedures used, melamine and glyoxylic acid were provided in the same and in a similar molar ratio as the 1 : 2.03 molar ratio used in Example I for melamine and methyl glyoxylate. In

² Accordingly, melamine and methyl glyoxylate were mixed in a molar ratio of 1 : 2.03, providing the following concentrations (weight %): 23.3% melamine and 33.0% methyl glyoxylate.

³ Although the specification recites "70° C", this is an obvious error as it should recite -70° C. One skilled in the art would have understood that the glass transition temperature of the washed powder including paraffin oil would be -70° C rather than +70° C.

Attempts A-C, the same molar ratio was used; in Attempt D, a molar ratio of 1 : 2.01 was used. Attempts A-D were performed as provided below.

Attempt A:

15. In a reaction vessel 25.03 gram melamine and 59.56 gram 50% glyoxylic acid were added. Melamine (100%) and glyoxylic acid (50%) were mixed in a molar ratio of 1 : 2.03, by providing the following concentrations (wt.%): 29.6% melamine and 35.2% glyoxylic acid. While at room temperature the mixture was a thick paste and could not be stirred. The mixture was heated in an oil bath at 80° C. Upon heating a stirrable mixture was obtained (at about 75° C). It was observed that the mixture started to gelate before it could turn into a clear solution; the mixture did not turn clear. Within about 7 minutes at 75 - 77° C the mixture became turbid and contained lumps. In the next 7 minutes (when the temperature reached about 82° C), the mixture turned white and became very thick. Mixing with an Ultra Turrax T25 was not possible.

Attempt B:

16. In a reaction vessel 59.59 gram glyoxylic acid (50%) was stirred with the Ultra Turrax T25 while being heated in an oil bath of about 80° C. While heating, 25.03 gram melamine (100%) was added gradually. The final molar ratio of melamine to glyoxylic acid was 1 : 2.03, by providing the following concentrations (in wt.%): 29.6% melamine and 35.2% glyoxylic acid. After a few minutes, however, the mixture was too thick to be mixed with the Ultra Turrax T25, even at 24000 rpm.

Attempt C:

17. The same approach was followed as in Attempt B, although when melamine was added, the glyoxylic acid was at room temperature. Before all of the melamine was added, the mixture turned so thick that it could no longer be stirred with the Ultra Turrax T25.

Attempt D:

18. To 25.05 gram melamine (100%), 59.10 gram glyoxylic acid (50%) was added in a reaction vessel during heating in an oil bath of 80° C. The molar ratio of melamine to glyoxylic acid was 1 : 2.01, by providing the following concentrations (in wt.%): 29.8% melamine and 35.1% glyoxylic acid. The mixture was stirred for 5 minutes while in the oil bath at 80° C. The mixture appeared to be a slurry, although it was not turbid as in Attempt A; the mixture did not turn clear. The reaction vessel (a 100 ml Schott-Duran bottle) was then placed under the Ultra Turrax T25 (while in an oil bath of about 50°C). While mixing with the Ultra Turrax T25 at 24000 RPM, paraffin oil colored with 0.1% Synthren Blue R obtained from Clariant (which is equivalent to Solvent Blue 122, a colorant similar to the one used in Example I of the present specification, but insoluble in hexane^{4/}) was added with a pipette in 3 minutes, during which time the mixture thickened relatively quickly. Within the next 7 minutes some Disperbyk-181 was added and then the mixture was poured into cold water (under gentle stirring).

19. A light blue slurry resulted. After stirring for one more hour (cooled in an ice bath), an attempt was made to filter the mixture over a Büchner funnel with

^{4/} The colorant used in Example I was not available when performing Attempts A-D.

filter paper. The filter became blocked immediately, however. The filtrate was significantly colored (light blue).

20. A small portion of the thicker part of the sediment was taken from the filter and dried during a weekend at 50° C under vacuum. The dried material was "greasy". The dried material was stirred in hexane, but the solvent did not become colored.^{5/} When alternatively the dried material was stirred in acetone, the acetone turned blue and the material lost some color intensity.

21. Further detail regarding Attempt D can be found in Tables I and II of the Appendix. In Table I the concentration percentages of melamine and glyoxylic acid are expressed as percentages based on the total weight of the composition, including paraffin oil and Polysynthren Blue R. Table II provides the concentration percentages by weight of melamine and glyoxylic acid corrected for the presence of the other ingredients. The concentrations (in wt.%) used in Attempt D, i.e., 29.8% melamine and 35.1% glyoxylic acid, are in accordance with Table II.

22. In all of the Attempts, problems were encountered. Obtaining a homogeneous melamine/glyoxylic acid mixture, which would be indicated by a clear solution, is important to attain proper encapsulation. In Attempt A, gelation began well before the solution had a chance to turn clear. The mixture became turbid and thick and could not be mixed with an Ultra Turrax T25. Because Attempt A was not successful in producing capsules, different yet analogous conditions were tried in Attempts B-D in which one or more parameters were changed to try to identify experimental parameters that may be suitable to obtain capsules. In these Attempts,

^{5/} Also after grinding the material no coloration of hexane was observed.

the order of addition of the various components was changed (Attempt B), the method of heating to the desired temperature was changed (Attempt C), and the molar ratio of melamine to glyoxylic acid was changed (Attempt D). In all of the Attempts, the mixtures obtained became too thick to be stirred with an Ultra Turrax T25.

23. Considerable thickening of the mixtures occurred in Attempts A, B, and C, and no capsules resulted from Attempts A, B, or C.

24. In Attempt D, a (non-turbid) slurry resulted from the reaction, yet no clear solution was observed. Thus, a homogeneous solution did not result. Although the mixture including the added coloring agent thickened to some extent, upon addition of a dispersing agent, it was possible to pour the mixture and place it in cold water (under gentle stirring). Upon filtering, however, the filter became blocked and the filtrate was significantly colored. The intended core material, the colorant, was observed in the filtrate. Therefore, encapsulation did not occur or sufficient encapsulation did not occur during the procedure of Attempt D.⁶

25. It is my opinion, in view of the results of Attempts A-D, that the reaction of melamine with glyoxylic acid does not result in sufficient encapsulation. In Attempts A-C, encapsulation did not result; in Attempt D, no capsules were produced or sufficient encapsulation did not result.

26. Because the encapsulation according to Example 1 of the present application which reacts melamine with methyl glyoxylate was found to produce sufficient encapsulation and Attempts A-D disclosed herein which react melamine with

⁶ The colorant used in Attempt D may be less suitable than the colorant in Example I of the present specification (because the colorant used in Attempt D does not dissolve sufficiently in an apolar medium). Yet the encapsulation was shown to be insufficient, regardless of the colorant used.

glyoxylic acid under similar experimental conditions did not produce sufficient encapsulation, it is my opinion that glyoxylic acid is not equivalent to methyl glyoxylate in its reaction with melamine in attempting to produce capsules.

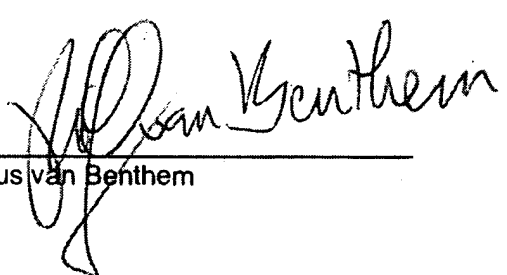
27. As noted above, Skoultchi discloses that either of the aldehydes, glyoxylic acid or an ester acetal of glyoxylic acid, can be reacted with a 2-imidazolidinone to prepare adducts useful as permanent press agents. One skilled in the art would have understood that Skoultchi discloses that the two aldehydes, namely the glyoxylic acid and the ester acetal of glyoxylic acid, are interchangeable in the reactions disclosed by Skoultchi to prepare permanent press agents. Yet it is shown here that glyoxylic acid is not equivalent to methyl glyoxylate in its reaction with melamine in attempting to produce capsules. The aldehydes disclosed in Skoultchi would not have been considered interchangeable with the aldehyde of Rätzsch with any expectation of success in preparing capsules. Thus, what one skilled in the art would have known – that the disclosure of Skoultchi regarding the preparation of adducts (using either of the aldehydes disclosed as equivalent, glyoxylic acid or methyl glyoxylate) for use as permanent press agents would not have provided any expectation of success in preparing capsules, as in the claimed process - has been confirmed.

28. In view of all of the foregoing, it is my opinion that one of ordinary skill in the art would not have expected success in the use of methyl glyoxylate and melamine in the claimed process for forming capsules in view of Rätzsch's disclosure of a melamine-aldehyde in the formation of capsules combined with Skoultchi's disclosure of 2-imidazolidinone reacted with glyoxylic acid or methyl glyoxylate for use as permanent press agents.

29. In view of all of the foregoing, it is also my opinion that the Examiner's alleged applicability of Skoultchi's disclosure of preparing permanent press agents in combination with Rätzsch for achieving the claimed process of making capsules in relation to the claimed process for forming capsules has been undermined.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 21 day of April 2010.



Rudolfus van Benthem

TABLE I

| Raw materials | M | Purity/ assay | d | g | g% | moles | mole ratio | vol. | conc. (wt%) | water (g) | wt. % against paraffin | wt. % against water |
|---------------------------|--------|------------------|------|--------|---------------|--------|---------------|------|----------------|--------------|------------------------------|---------------------------|
| Melamine | 126.12 | 100.0% | 1.57 | 25.05 | 25.05 | 0.1986 | 1.000 | 16.0 | 23.4% | | | |
| Glyoxylic acid | 74.03 | 50.0% | 1.3 | 59.10 | 29.55 | 0.3992 | 2.010 | 45.5 | 27.5% | 29.55 | | |
| Water | 18 | 100.0% | 1 | 0.00 | 0.00 | 0.0000 | 0.000 | 0.0 | 0.0% | | | |
| Paraffin oil | 352.00 | 100.0% | 0.86 | 21.80 | 21.80 | 0.0619 | 0.312 | 25.3 | 20.3 | | | |
| Polysynthren Blue R | 372.37 | 100.0% | 1.45 | 0.0220 | 0.0220 | 0.0001 | 0.000 | 0.0 | 0.02% | | 0.1009% | |
| Disperbyk-181 | 0 | 100.0% | 1.04 | 1.30 | 1.30 | | | 1.3 | 1.2% | | | 4.40% |
| total mass: | | | | 107.27 | total volume: | | | | 88.0 | 29.55 | | |
| Ratio paraffin oil/water: | | | | 0.74 | | | | | | | | |

TABLE II

| Raw materials | M | Purity/ assay | d | g | g% | moles | mole ratio | vol. | conc. (wt%) | water (g) | wt.% against paraffin | wt.% against water |
|---------------------------|--------|------------------|------|--------|---------------|--------|---------------|------|----------------|--------------|-----------------------------|--------------------------|
| Melamine | 126.12 | 100.0% | 1.57 | 25.05 | 25.05 | 0.1986 | 1.000 | 16.0 | 29.8% | | | |
| Glyoxylic acid | 74.03 | 50.0% | 1.3 | 59.10 | 29.55 | 0.3992 | 2.010 | 45.5 | 35.1% | 29.55 | | |
| Water | 18 | 100.0% | 1 | 0.00 | 0.00 | 0.0000 | 0.000 | 0.0 | 0.0% | | | |
| Paraffin oil | 352.00 | 100.0% | 0.86 | 0.00 | 0.00 | 0.0000 | 0.000 | 0.0 | 0.0% | | | |
| Polysynthren Blue R | 372.37 | 100.0% | 1.45 | 0.0000 | 0.0000 | 0.0000 | 0.000 | 0.0 | 0.00% | | | |
| Disperbyk-181 | 0 | 100.0% | 1.04 | 0.00 | 0.00 | | | 0.0 | 0.0% | | | 0.00% |
| total mass: | | | | 84.15 | total volume: | | | | 61.4 | 29.55 | | |
| Ratio paraffin oil/water: | | | | 0.00 | | | | | | | | |

M = Molecular mass
d = density (g/cm³)
g = grams

g% = grams correcting for purity
vol. = volume (cm³)

EXHIBIT B

Curriculum Vitae Prof. Dr R.A.T.M. van Benthem

1. Personal

Family name: Van Benthem
First name: Rolf
First names (full): Rudolfus Antonius Theodorus Maria
Date of birth: March 21, 1968
Place of birth: Roosendaal en Nispen
Marital state: Married, 2 children

Address: Sportlaan 9, 6141 BR Limbricht, The Netherlands
tel: +31 46 4589348
e-mail (private): r.van.benthem@hetnet.nl

2. Education

Highschool: Gymnasium-B (27-05-86, *Cum Laude*)
Gertrudis Lyceum, Roosendaal

Masters degree (MSc): Scheikunde (Chemistry), 1986-1990 (22-12-90, *Cum Laude*), Leiden University
Research subject (Prof. Dr. A. Van der Gen): Biocatalysis, organic synthesis

Thesis (PhD): AIO (01-01-91 /31-12-94), promotion date 22-04-95
University of Amsterdam
Promotor Prof. Dr. W.N. Speckamp
Copromotor Prof. Dr. H. Hiemstra
Research: Homogeneous catalysis, organic synthesis.
Thesis title: "*Palladium Cluster Catalyzed Oxidative Cyclizations in the Synthesis of Aminoalkenols and Diamines*"

Awards / honors: Received Dr. H. J. Backer award 1995 (26-04-96) (*best Dutch thesis in organic chemistry 1995*)

3. Profesional career

DSM: May 1, 1995 to present.

Current position: Competence Manager Network Chemistry
Competence area Chemistry of Resins and Polymers

This concerns a worldwide responsibility of management) of DSM's knowledge (ensure access to, secure quality of, gap analysis and gap closure strategy of) in thermoset materials, a.o. coatings, composites, rubbers and adhesives, and crosslinking agents.

| | |
|---------------------|--|
| Department | DSM Ahead BV, PM-CT-Thermosets DSM Engineering Plastics |
| Previous positions | Senior Scientist Coating Resins (2000-2002) Program Manager Coating Resins (1999-2005) Project leader Coating Resins (1995-2005) Project leader Fundamental Melamine Resins Research (1998-2005) Group leader Resins (1998-2005) |
| Awards / honors | DSM Publication Award 2001 <i>"Synthesis and Characterization of Bis(2-hydroxy-propyl)amide based Hyperbranched Polyesterarnides"</i> R.A.T.M. van Benthem, E. Gelad, N. Meijerink, P. Froehling, P.H.M. Hendriks, D. Muscat, T.I.G. Zwartkruis, E.G. de Koster, <i>Macromolecules</i> 2001 34(11), 3559-3566. DSM Research Award 2004 |
| Address | Research Campus Geleen PO Box 18, 6160 MD Geleen, The Netherlands tel +31 46 47 67052 fax +31 46 47 63949 e-mail: rolf.benthem-van@dsm.com |
| TU-Eindhoven | October 1, 1992 to present |
| Position | Professor of Coatings Technology Appointed in part-time (0.2 FTE) Laboratory of Material and Interface Chemistry (SMG) Department of Chemical Engineering (ST) |
| Address | Eindhoven University of Technology Den Dolech 2 PO Box 5600 MB Eindhoven tel +31 40 247 2029 / 2770 fax +31 40 247 5619 e-mail: r.a.t.m.v.benthem@tue.nl |

Other

Chairman Coatings Science International (www.coatings-science.com, 2005 – present)

Member Matiello Lecture Selection Committee (2006 – 2011), Federation of Societies for Coatings Technology, (FCST).

Member Advisory Committee IOP Selfhealing Materials (2006-2009).

4. Publications and Patents

SciFinder June 2009: 83 publications
48 published patents (of which 42 filed by DSM)

Recent publications:

Journals

2010

With, G. de, Benthem, R.A.T.M. van (2010). Editorial POC CoSi 2009. *Progress in Organic Coatings*, submitted / in press.

Senatore, D., Laven, J., Benthem, R.A.T.M. van, Camera, D. la, With, G. de (2010). Microencapsulation of Epoxidized Linseed Oil Liquid Cross-Linker in Poly(N-vinylpyrrolidone): Optimization by a Design-of-Experiments Approach. *Industrial and Engineering Chemistry Research*, 49(8), 3642-3653.

Foyet, A., Wu, T., Ven, L.G.J. van der, With, G. de, Benthem, R.A.T.M. van (2010). Absorption of water and corrosion performance of a clear and pigmented epoxy coating on Al-2024 alloy. *ECS Transactions*, 25(29), 31-39.

2009

With, G. de, Benthem, R.A.T.M. van (2009). Editorial POC CoSi 2008. *Progress in Organic Coatings*, 64(2-3), 109-111.

Soer, W.J., Ming, W., Koning, C.E., Benthem, R.A.T.M. van, Mol, J.M.C., Terryn, H. (2009). Barrier and adhesion properties of anti-corrosion coatings based on surfactant-free latexes from anhydride-containing polymers. *Progress in Organic Coatings*, 65(1), 94-103.

Noordover, B.A.J., Heise, A., Malanowski, P., Senatore, D., Mak, M.H.A.P., Molhoek, L.J., Duchateau, R., Koning, C.E., Benthem, R.A.T.M. van (2009). Biobased step-growth polymers in powder coating applications. *Progress in Organic Coatings*, 65(2), 187-196.

Malanowski, P., Huijser, S., Scaltro, F., Benthem, R.A.T.M. van, Ven, L.G.J. van der, Laven, J., With, G. de (2009). Molecular mechanism of photolysis and photooxidation of poly(neopentyl isophthalate). *Polymer*, 50(6), 1358-1368.

Malanowski, P., Huijser, S., Benthem, R.A.T.M. van, Ven, L.G.J. van der, Laven, J., With, G. de (2009). Photodegradation of poly(neopentyl isophthalate) part I: Laboratory

and outdoor conditions. *Polymer Degradation and Stability*, 94(11), 2086-2094.

Foyet, A., Wu, T., Kodentsov, A., Ven, L.G.J. van der, With, G. de, Benthem, R.A.T.M. van (2009). Impedance evaluation of permeability and corrosion of Al-2024 aluminum alloy coated with a chromate free primer. *Progress in Organic Coatings*, 65(2), 257-262.

Foyet, A., Wu, T., Ven, L.G.J. van der, Kodentsov, A., With, G. de, Benthem, R.A.T.M. van (2009). Influence of mixing ratio on the permeability of water and the corrosion performance of epoxy/amine coated un-pretreated Al-2024 evaluated by impedance spectroscopy. *Progress in Organic Coatings*, 64(2-3), 138-141.

Wietor, J.L., Dimopoulos, A., Govaert, L.E., Benthem, R.A.T.M. van, With, G. de, Sijbesma, R.P. (2009). Preemptive healing through supramolecular cross-links. *Macromolecules*, 42(17), 6640-6646.

2008

With, G. de, Benthem, R.A.T.M. van (2008). Editorial POC CoSi 2007. *Progress in Organic Coatings*, 61(2-4), 103-105.

Soer, W.J., Ming, W., Koning, C.E., Benthem, R.A.T.M. van (2008). Towards anti-corrosion coatings from surfactant-free latexes based on maleic anhydride containing polymers. *Progress in Organic Coatings*, 61(2-4), 224-232.

Soer, W.J., Ming, W., Koning, C.E., Benthem, R.A.T.M. van (2008). Crosslinking systems and film properties for surfactant-free latexes based on anhydride-containing films. *Polymer*, 49(16), 3399-3412.

Wu, D., Ming, W., Benthem, R.A.T.M. van, With, G. de (2008). Superhydrophobic fluorinated polyurethane films. *Journal of Adhesion Science and Technology*, 22(15), 1869-1881.

Senatore, D., Berix, M.J.A., Laven, J., Benthem, R.A.T.M. van, With, G. de, Mezari, B., Magusin, P.C.M.M. (2008). Miscibility and specific interactions in blends of poly(n-vinyl-2-pyrrolidone) and acid functional polyester resins. *Macromolecules*, 41(21), 8020-8029.

Dikic, T., Ming, W., Thüne, P.C., Benthem, R.A.T.M. van, With, G. de (2008). Well-defined polycaprolactone precursors for low surface-energy polyurethane films. *Journal of Polymer Science, Part A: Polymer Chemistry*, 46(1), 218-227.

Wijting, W.K., Laven, J., Benthem, R.A.T.M. van, With, G. de (2008). Adsorption of ethoxylated styrene oxide and polyacrylic acid and mixtures thereof on organic pigment. *Journal of Colloid and Interface Science*, 327(1), 1-8.

2007

With, G. de, Benthem, R.A.T.M. van (2007). Editorial - Progress in Organic Coatings, vol. 58, no. 2-3 (2007). *Progress in Organic Coatings*, 58(2-3), 77-79.

With, G. de, Benthem, R.A.T.M. van (2007). Progress in Organic Coatings, editorial, vol. 58, issues 2-3. *Progress in Organic Coatings*, 58(2-3), 77-79.

Noordover, B.A.J., Duchateau, R., Benthem, R.A.T.M. van, Ming, W., Koning, C.E. (2007). Enhancing the functionality of biobased polyester coating resins through

modification with citric acid. *Biomacromolecules*, 8(12), 3860-3870.

Wu, D., Vrancken, R.J., Loenen, B.G.H. van, Benthem, R.A.T.M. van, With, G. de, Ming, W. (2007). Lipophobicity on hierarchically structured superhydrophobic surfaces. *Polymeric Materials: Science and Engineering*, 97, 418-419.

Senatore, D., Cate, A.T. ten, Laven, J., Benthem, R.A.T.M. van, With, G. de (2007). Controlled release of micro-encapsulated cross-linker in powder coatings. *PMSE Preprints*, 97, 912-913.

Dikic, T., Erich, S.J.F., Ming, W., Huinink, H.P., Thüne, P.C., Benthem, R.A.T.M. van, With, G. de (2007). Fluorine depth profiling by high-resolution 1D magnetic resonance imaging. *Polymer*, 48(14), 4063-4067.

Haveren, J. van, Oostveen, E., Micciche, F., Noordover, B.A.J., Koning, C.E., Benthem, R.A.T.M. van, Frissen, A.E., Weijnen, J.G.J. (2007). Resins and additives for powder coatings and alkyd paints, based on renewable resources. *Journal of Coatings Technology and Research*, 4(2), 177-186.

2006

With, G. de, Benthem, R.A.T.M. van (2006). Editorial: Coatings Science International Conference 2005. *Progress in Organic Coatings*, 55(2), 71-74.

Soer, W.J., Ming, W., Klumperman, L., Koning, C.E., Benthem, R.A.T.M. van (2006). Surfactant-free artificial latexes from modified styrene-maleic anhydride (SMA) copolymers. *Polymer*, 47(22), 7621-7627.

Noordover, B.A.J., Staaldin, V.G. van, Duchateau, R., Koning, C.E., Benthem, R.A.T.M. van, Mak, M.H.A.P., Heise, A., Frissen, A.E., Haveren, J. van (2006). Co- and terpolyesters based on isosorbide and succinic acid for coating applications: Synthesis and Characterization. *Biomacromolecules*, 7(12), 3406-3416.

2005

Ming, W., Wu, D., Benthem, R.A.T.M. van, With, G. de (2005). Biomimetic creation of superhydrophobic films. *PMSE Preprints*, 93, 259.

Ming, W., Wu, D., Benthem, R.A.T.M. van, With, G. de (2005). Superhydrophobic Films from Raspberry-like Particles. *Nano Letters*, 5(11), 2298-2301.

2004

Schmatloch, S., Bach, H., Benthem, R.A.T.M. van, Schubert, U.S. (2004). High-throughput experimentation in organic coating and thin film research : state-of-the-art and future perspectives. *Macromolecular Rapid Communications*, 25(1), 95-107.

2003

Benthem, R.A.T.M. van, Hofland, A., Peerlings, H.W.I., Meijer, E.W. (2003). Ideally selective diisocyanate building blocks New perspectives for dendrimers and coating binders. *Progress in Organic Coatings*, 48(2-4), 164-176.

2001

Peerlings, H.W.I., Benthem, R.A.T.M. van, Meijer, E.W. (2001). Fast and convenient

construction of carbamate/urea-based dendrimers by making use of a diisocyanate building block. *Journal of Polymer Science, Part A: Polymer Chemistry*, 39(18), 3112-3120.



Book (chapters)

2009

Wu, D., Ming, W., Benthem, R.A.T.M. van, With, G. de (2009). Superhydrophobic fluorinated polyurethane films. In A. Carré, K.L. Mittal (Eds.), *Superhydrophobic Surfaces*. (pp. 139-151) Leiden: Brill.

2007

Benthem, R.A.T.M. van, Ming, W., With, G. de (2007). Self healing polymer coatings. In S. van der Zwaag (Ed.), *Self healing materials : an alternative approach to 20 centuries of materials science*. (pp. 139-159) Berlin: Springer.

2005

Benthem, R.A.T.M. van, Evers, L.J., Mattheij, J., Hofland, A., Molhoek, L.J., Koning, A.J. de, Jansen, J.F.G.A., Duin, M. van (2005). Thermosets. In T. Meyer, J.T.F. Keurentjes (Eds.), *Handbook on polymer reaction engineering*. (pp. 833-910) Wiley-VCH.



Congress, Symposia

2008

Dikic, T., Ming, W., Thüne, P.C., Tian, M., Benthem, R.A.T.M. van, With, G. de (2008). Self-replenishing low surface-energy polyurethane films. *Abstracts of Papers, 235th ACS National Meeting, New Orleans, LA, United States*. (pp. POLY-612). Washington, D. C: American Chemical Society.

2007

Noordover, B.A.J., Duchateau, R., Koning, C.E., Benthem, R.A.T.M. van, Ming, W., Haveren, J. van, Es, D.S. van (2007). Biobased functional polyesters for coating applications: Synthesis, characterization and application. *Abstracts of Papers of the 233rd ACS National Meeting, 25-29 March 2007, Chicago, Illinois, USA*. (pp. CELL-172).

Senatore, D., Cate, T.A. ten, Laven, J., Benthem, R.A.T.M. van, With, G. de (2007). Controlled release of micro-encapsulated cross-linker in powder coatings. *Abstracts of Papers, 234th ACS National Meeting, Boston, MA, United States, August 19-23, 2007*. (pp. PMSE-517).

Dikic, T., Ming, W., Benthem, R.A.T.M. van, With, G. de (2007). Self-replenishing low-adherence coatings. In A.J.M. Schmets, S. van der Zwaag (Eds.), *Proceedings of the 1st International Conference on Self Healing Materials, 18-20 April 2007, Noordwijk, The Netherlands*. Springer.

2005

Ming, W., Wu, D., Benthem, R.A.T.M. van, With, G. de (2005). Biomimetic creation of superhydrophobic films. *Abstracts of papers, 230th ACS national meeting : August 28 - September 1, 2005, Washington, DC. (pp. PMSE-165)*. Washington, United States, : American Chemical Society.

Dikic, T., Ming, W., Benthem, R.A.T.M. van, With, G. de (2005). Synthesis of well-defined perfluoroalkyl-end-capped polymers by ring-opening polymerization. *International Conference on Fluorine and Silicone Coatings. (pp. Paper-3)*. United Kingdom, Manchester.